LAWS OF LARGE NUMBERS FOR MESOSCOPIC STOCHASTIC MODELS OF REACTING AND DIFFUSING PARTICLES

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ABSTRACT. We study the asymptotic behaviour of some mesoscopic stochastic models for systems of reacting and diffusing particles (also known as density-dependent population processes) as the number of particles goes to infinity. Our approach is related to the variational approach to solving the parabolic partial differential equations that arise as limit dynamics. We first present a result for a model that converges to a classical system of reaction-diffusion equations. In addition, we discuss two models with nonlinear diffusion that give rise to quasilinear parabolic equations in the limit.

Key words: interacting random processes, reaction-diffusion model, law of large numbers

1. Introduction

In this paper we study the asymptotic behaviour of certain mesoscopic stochastic particle models (or density-dependent population processes) for reaction-diffusion systems as the number of particles goes to infinity. Mesoscopic stochastic particle models are informally defined as follows. We think of a chemical reactor as being composed of cells or compartments of mesoscopic size l. Each cell may contain up to about n particles of each species. Particles of species j jump randomly from a cell to an adjacent one in direction $\pm e_k \in \mathbb{R}^m$ according to rates $d_{j,k\pm}$ which may be functions of the particle densities in the cell (the particle numbers divided by n) and their discrete gradients. Moreover, if we denote the vector of particle densities in cell z at time t by $\boldsymbol{u}_l(z,t) = (u_{l,1}(z,t), \ldots, u_{l,n_s}(z,t))$, n_s being the number of species, then the number of particles in cell z changes randomly with rate $n\, K_i(\boldsymbol{u}_l(z,t))$ according to the stoichiometry of the ith reaction, $i = 1, \ldots, n_r$. Thus the model can, in the simplest case, be thought of as a combination of a continuous-time version of the classical urn model by P. and T. Ehrenfest for diffusion through a membrane and the standard stochastic model for chemical reactions (van Kampen, 1992). We call this type of model mesoscopic because interactions between individual particles are not taken into account explicitly.

Stochastic particle models of this type have been described and studied by many authors in physics (Nicolis & Prigogine, 1977; Gillespie, 1977; Haken, 1983; van Kampen, 1992; Gardiner, 2004) and mathematics (Kurtz, 1977/78, 1981; Arnold & Theodosopulu, 1980; Kotelenez, 1986, 1988; Blount, 1991, 1993, 1994; Guiaş, 2002; Ball et al., 2006). In the physical literature the model is often simply called 'the' stochastic model for chemical reactions.

Our aim is to derive partial differential equations (PDEs) as macroscopic limit equations for $l \to 0$, $n \to \infty$ with $d_{j,k\pm}$ suitably adjusted. To this end, we generally proceed in two steps. We first study the convergence of a semi-discrete finite-difference approximation of the limit equations where the spatial derivatives are replaced by finite differences. Having established the convergence of the semi-discrete approximation, the second step in the proofs consists in estimating the distance between the approximation and the particle densities associated to the stochastic particle model in an appropriate norm. This procedure is motivated by the observation that the particle densities generally satisfy a system of stochastic differential equations that can be regarded as a spatially semi-discretised finite-difference approximation of the macroscopic PDEs perturbed by a martingale noise term. In previous work (Kotelenez, 1986, 1988; Blount, 1991, 1993, 1994; Guiaş, 2002) laws of large numbers have been shown for linear and certain nonlinear models by means of semigroup methods. In particular, the solutions of the limit equations have been characterised as the mild solutions that one obtains from the semigroup approach to linear and semilinear parabolic equations. Our method is related to the variational approach to parabolic PDEs. The solution of the limit equation is an appropriately defined weak solution the existence of which can be established with Hilbert-space methods.

The paper is organised as follows. In the next section we introduce the macroscopic PDE model and the mesoscopic stochastic particle model in their most general form. In Section 3 we describe the results for three particular instances of the general models. We first consider a stochastic model leading to a classical system of reaction-diffusion equations as limit dynamics. Subsequently, we discuss two models with a nonlinear diffusion mechanism. For the sake of simplicity, we restrict the discussion to a single-species model without chemical reactions. In Section 3.2 we investigate what happens when the intensity for a jump of a particle to a neighboring cell depends on the local concentration, i.e., $d_{k\pm}(u_l(z), \pm\nabla^{\pm}u_l(z)) = d(u_l(z))/(2m)$, where d is monotonously increasing. Thereafter, in Section 3.3, we have a look at an example where the intensity for a jump to a neighboring cell depends on the absolute value of the (discrete) concentration gradient, i.e., $d_{k+}(u_l(z), \nabla^{+}u_l(z)) = d(\partial_k^{+}u_l(z))$ for a jump to the right and $d_{k-}(u_l(z), -\nabla^{-}u_l(z)) = d(-\partial_k^{-}u_l(z))$ for a jump to the left, respectively, for a monotonously increasing and symmetric function d. (See below for the definition of $\partial_k^{\pm}$.) Nonconstant diffusion coefficients play a role in the modelling of self-organisation of microorganisms (Ben-Jacob et al., 2000) and surface reactions (Naumovets, 2005). Finally, in Section 5 the results are discussed and related to other work.

2. The general models

A basic verbal description of a chemical reactor is given in terms of its geometry and a system of chemical equations for the reaction under consideration:

$$
\begin{aligned}
0+ n_{1,1}\ \mathrm{C}_1 + \cdots + n_{1,n_s}\ \mathrm{C}_{n_s} &\rightarrow 0+ \tilde n_{1,1}\ \mathrm{C}_1 + \cdots + \tilde n_{1,n_s}\ \mathrm{C}_{n_s} \\
&\vdots \\
0+ n_{n_r,1}\ \mathrm{C}_1 + \cdots + n_{n_r,n_s}\ \mathrm{C}_{n_s} &\rightarrow 0+ \tilde n_{n_r,1}\ \mathrm{C}_1 + \cdots + \tilde n_{n_r,n_s}\ \mathrm{C}_{n_s}.
\end{aligned} \tag{1}
$$

Here $n_s \in \mathbb{N}$ denotes the number of different species present in the reactor, $n_r \in \mathbb{N}$ the number of reactions and $n_{ij}, \tilde n_{ij} \in \mathbb{Z}$, $i = 1, \dots, n_r$, $j = 1, \dots, n_s$, are the stoichiometric coefficients. All particles coming from or going to one or several reservoirs coupled to the reactor are denoted by '0'. Note that we count reverse reactions separately. The term 'chemical reaction' is understood in a broad sense, i.e., the reactions under consideration are not supposed to be 'elementary reactions' in a dilute solution. The geometry of the chemical reactor is represented by a bounded domain $G \subset \mathbb{R}^m$, $m = 1, 2, 3$, with Lipschitz boundary. We generally assume that mass transfer in the reactor occurs only by diffusion. In addition, we take into account inflow and outflow of mass from and to the reservoirs.

2.1. **The general macroscopic model.**

On the macroscopic level the dynamics of the densities u_j of the chemical species C_j is described by a system of n_s mass-balance equations in the space-time domain $Q_T = G \times (0, T)$, $T > 0$ being the time of observation:

$$
\partial_t u_j + \nabla \cdot \boldsymbol{J}_j(x, \boldsymbol{u}, \nabla \boldsymbol{u}) = f_j(x, \boldsymbol{u}), \quad j = 1, \dots, n_s. \tag{2}
$$

Here $\boldsymbol{u} = (u_1, \dots, u_{n_s})$, and $\nabla \boldsymbol{u} = \big((\nabla u_1)^T, \dots, (\nabla u_{n_s})^T\big)^T$. The vector-valued functions $\boldsymbol{J}_j : \mathbb{R}^m \times \mathbb{R}^{n_s} \times \mathbb{R}^{n_s \times m} \rightarrow \mathbb{R}^m$ are appropriate 'constitutive laws' for the diffusive mass flux and the functions $f_j : \mathbb{R}^m \times \mathbb{R}^{n_s} \rightarrow \mathbb{R}$ describe the contributions of the chemical reactions. In addition, appropriate boundary and initial conditions have to be specified. In the particular instances of Eq. (2) considered below we assume that the reaction functions f_j and the fluxes $\boldsymbol{J}_j$ do not depend explicitly on the space variable x. The reaction functions f_j are obtained in the following way. We assume that the density u_j of the jth species changes with rate $\nu_{ij} K_i(\boldsymbol{u})$ due to the ith reaction, where the reaction rates $K_i : \mathbb{R}^{n_s} \rightarrow \mathbb{R}$, $i = 1, \dots, n_r$, are functions of the local particle densities, and the matrix $(\nu_{ij}) \in \mathbb{Z}^{n_r \times n_s}$ is defined by $\nu_{ij} = \tilde n_{ij} - n_{ij}$. Then

$$
f_j(\boldsymbol{u}) = \sum_{i=1}^{n_r} \nu_{ij} K_i(\boldsymbol{u}). \tag{3}
$$

Unfortunately, there is no unified existence theory of Eq. (2). The notions of solution for the particular instances of Eq. (2) that will appear as limit dynamics of stochastic particle models are discussed below.

2.2. **The general mesoscopic stochastic particle model.** To motivate the set-up of our model we briefly discuss the characteristic time and length scales in a reaction-diffusion system. In a reaction-diffusion system typically three different characteristic length scales can be identified: the total size of the system L, a 'diffusion length' l, which corresponds to the size of a well-mixed cell or compartment, and the typical distance of a particle to its nearest neighbour λ. We postulate

$$\lambda << l << L,$$

which is certainly a reasonable assumption for many systems. The micro-scale λ will not appear explicitly in the mesoscopic model. These three length scales lead in a natural way to two ratios,

$$N = L/l >> 1 \quad \text{and} \quad n = l/\lambda >> 1,$$

which in one space dimension correspond to the number of cells and the typical number of particles per cell (or sites per cell, if we think of the particles as being located at the points of a sublattice), respectively. The law of large numbers we are aiming at can be regarded as an idealisation obtained by letting both ratios tend to infinity. In our approach we keep the system size L fixed. Hence, the cell size l and the typical inter-particle distance λ must go to zero. Alternatively, we could fix λ and let l and L tend to infinity.

In a similar way one can identify three different time scales: a time scale which corresponds to the time needed by a particle to travel the distance λ (or a hopping rate from site to site δ) and does not appear explicitly in the mesoscopic model, a time scale which corresponds to the 'hopping rate' d from cell to cell, and, finally, the time of observation T. We assume

$$1/\delta << 1/d << T,$$

so that a cell can always be regarded as well-mixed. As for the chemical reactions, we assume that the typical time between two reaction events in a cell is of order $1/n$.

We now introduce the state space of the stochastic particle model. It will turn out to be useful to regard the stochastic particle densities as elements of a discrete version of the Lebesgue space $L^2(G)$. Discrete Lebesgue spaces are used in numerical analysis and are defined, e.g., in Zeidler (1990). For the convenience of the reader we repeat the definition here. We first choose a cubic lattice in $\mathbb{R}^m$ with grid mesh $h \in I = (0, h_0] \subset \mathbb{R}^+$. More precisely, for some fixed $z_0 \in \mathbb{R}^m$ we define the set of vertices $\mathcal{Z}_h(z_0)$ by

$$\mathcal{Z}_h(z_0) = \Big\{ z \in \mathbb{R}^m : z = hz_0 + \sum_{k=1}^{m} i_k h\, e_k,\ i_1, \ldots, i_m \in \mathbb{Z} \Big\}, \tag{4}$$

where e_k denotes the kth unit vector in $\mathbb{R}^m$. The kth coordinate of a vertex is thus an integer multiple of h shifted by $h\, z_{0,k}$. To each vertex $z \in \mathcal{Z}_h(z_0)$

we assign an open cube $c_h(z) \subset \mathbb{R}^m$ with edges parallel to the coordinate axis having edge length h and z as midpoint.

Definition 2.1. The set $\mathcal{G}_h$ of *interior lattice points* of the domain G generated by the lattice $\mathcal{Z}_h(z_0)$ is defined as

$$\mathcal{G}_h = \big\{z \in \mathcal{Z}_h(z_0) : c_h(z) \subset G\big\}.$$

Definition 2.2. By a *lattice function* we understand a function $u_h : \mathcal{Z}_h(z_0) \to \mathbb{R}$, i.e., a function that assigns a real number to each vertex $z \in \mathcal{Z}_h(z_0)$. The *extended version* of a lattice function is the step function $\tilde{u}_h : \mathbb{R}^m \to \mathbb{R}$, $x \mapsto \sum_{z\in\mathcal{Z}_h(z_0)} u_h(z)\, \mathbb{1}_{c_h(z)}(x)$, where $\mathbb{1}_{c_h(z)}$ is the indicator function of the open cube $c_h(z)$.

Definition 2.3. The *discrete Lebesgue space* $\mathcal{L}^2(\mathcal{G}_h)$ is the space of lattice functions that vanish outside $\mathcal{G}_h$ equipped with the scalar product

$$\big(u_h,\, v_h\big)_{\mathcal{L}^2(\mathcal{G}_h)} = h^m \sum_{z\in\mathcal{G}_h} u_h(z)\, v_h(z) = \int_{\mathbb{R}^m} \tilde{u}_h(x)\, \tilde{v}_h(x)\, dx.$$

For the sake of brevity, we usually skip the tilde notation and use the same symbol for u_h, $\tilde{u}_h$ and $\tilde{u}_h|_G$ if there is no risk of confusion.

We now identify the well-mixed cells in the chemical reactor represented by the domain G with the open cubes $c_l(z)$ around the interior lattice points $z \in \mathcal{G}_l$ generated by a grid $\mathcal{Z}_l(z_0)$. The state space S_l of the particle density process $\boldsymbol{u}_l(t) = (u_{l,1}(t), \dots, u_{l,n_s}(t))$, $t \geq 0$, to be described below is defined as the (countable) set of vector-valued lattice functions from the space $(\mathcal{L}^2(\mathcal{G}_l))^{n_s}$ that take values in the set $\frac{1}{n}\mathbb{N}_0^{n_s}$ endowed with the induced metric.

In order to characterise the random dynamics in the state space S_l, we still need the following definitions.

Definition 2.4. The set of lattice points $\mathcal{G}_h^1$ is defined as

$$\mathcal{G}_h^1 = \big\{z \in \mathcal{G}_h :\; z \pm h\, e_k \in \mathcal{G}_h,\; k = 1, \dots, m\big\}.$$

Definition 2.5. For a lattice function u_h the *discrete derivatives* $\partial_k^+ u_h$ and $\partial_k^- u_h$ are defined as the lattice functions given by

$$\partial_k^{\pm} u_h(z) = \frac{u_h(z \pm h\, e_k) - u_h(z)}{\pm h}, \quad k = 1 \dots, m.$$

Higher derivatives are obtained by repeated application of $\partial_k^{\pm}$.

Now let, for $z \in \mathcal{G}_l^1$ and $j = 1, \dots, n_s$, $\boldsymbol{\chi}_{j,z} \in S_l$ be the state with particle density one for species j in cell z and zero elsewhere. For $z \in \mathcal{G}_l \setminus \mathcal{G}_l^1$ we define $\boldsymbol{\chi}_{j,z}$ identically zero. The random dynamics of the particle densities is characterised by the following set of transition intensities $q_l(\cdot,\cdot)$ for jumps from a state $\boldsymbol{u}_l \in S_l$ to other states.

- A particle of species j may leave cell $z \in \mathcal{G}_l^1$ and jump to $z \pm l e_k$:

$$
\begin{aligned}
&q_l(\boldsymbol{u}_l, \boldsymbol{u}_l - \tfrac{1}{n}\boldsymbol{\chi}_{j,z} + \tfrac{1}{n}\boldsymbol{\chi}_{j,(z-le_k)}) = n\, d_{j,k-}(\boldsymbol{u}_l(z), -\nabla^- \boldsymbol{u}_l(z))\, u_{l,j}(z),\\
&q_l(\boldsymbol{u}_l, \boldsymbol{u}_l - \tfrac{1}{n}\boldsymbol{\chi}_{j,z} + \tfrac{1}{n}\boldsymbol{\chi}_{j,(z+le_k)}) = n\, d_{j,k+}(\boldsymbol{u}_l(z), \nabla^+ \boldsymbol{u}_l(z))\, u_{l,j}(z),
\end{aligned} \tag{5}
$$

where $d_{j,k\pm}(\cdot,\cdot) \geq 0$ is the hopping rate of species j in the direction $\pm e_k$, which may be a function of the local densities $u_{l,j}$ and their discrete gradients $\nabla^\pm u_{l,j} = (\partial_1^\pm u_{l,j}, \dots, \partial_m^\pm u_{l,j})$. Note that the particles vanish if they attempt to jump to a cell at the boundary, which corresponds to homogeneous Dirichlet boundary conditions.

- The number of particles in cell $z \in \mathcal{G}_l^1$ changes by reaction i:

$$
q_l(\boldsymbol{u}_l, \boldsymbol{u}_l + \tfrac{1}{n}\textstyle\sum_{j=1}^{n_s}\nu_{ij}\boldsymbol{\chi}_{j,z}) = n\, K_i(\boldsymbol{u}_l(z)) \quad \text{if } \boldsymbol{u}_l + \tfrac{1}{n}\textstyle\sum_{j=1}^{n_s}\nu_{ij}\boldsymbol{\chi}_{j,z} \in S_l. \tag{6}
$$

Here we use the same reaction rates as in the deterministic model. A slight generalisation could be obtained by adding lower order terms.

The intensity for other possible transitions is zero.

For simplicity we always assume that $\boldsymbol{u}_l(0) \in S_l$ is non-random. In all cases considered below the transition intensities $q(\cdot,\cdot)$ characterise a Markov jump process $(\boldsymbol{u}_l(t))_{t\geq 0}$ (with respect to the induced filtration) on some probability space $(\Omega, \mathscr{A}, P)$ with values in S_l starting at $\boldsymbol{u}_l(0)$ that corresponds to a Feller semigroup with generator L_l defined by

$$
L_l g(\boldsymbol{u}_l) = \sum_{\tilde{\boldsymbol{u}}_l \neq \boldsymbol{u}_l} q_l(\boldsymbol{u}_l, \tilde{\boldsymbol{u}}_l)\,\big(g(\tilde{\boldsymbol{u}}_l) - g(\boldsymbol{u}_l)\big), \quad g \in \hat{C}(S_l). \tag{7}
$$

Here $\hat{C}(S_l)$ denotes the space of bounded continuous functions from S_l to $\mathbb{R}$. (Note that continuity is trivial.) This follows from Theorem 3.1 in Chapter 8 of Ethier & Kurtz (1986).

For later use we now introduce a discrete version of the Sobolev space $H_0^1(G)$ (the subspace of functions in $L^2(G)$ that have weak partial derivatives in $L^2(G)$ and vanish on the boundary of G).

Definition 2.6. By the *discrete Sobolev space* $\mathcal{H}_0^1(\mathcal{G}_h)$ we understand the set of all lattice functions that vanish outside $\mathcal{G}_h^1$ equipped with the scalar product

$$
\big(u_h,\, v_h\big)_{\mathcal{H}_0^1(\mathcal{G}_h)} = \big(u_h,\, v_h\big)_{\mathcal{L}^2(\mathcal{G}_h)} + \sum_{k=1}^{m} \big(\partial_k^+ u_h,\, \partial_k^+ v_h\big)_{\mathcal{L}^2(\mathcal{G}_h)}.
$$

The space $\mathcal{H}_0^1(\mathcal{G}_h)$ has many properties in common with the Sobolev $H_0^1(G)$ defined on a continuous domain G, e.g., we have a discrete integration by parts formula and a discrete version of Poincaré's inequality. (Here and in the following C denotes a generic constant that may change from line to line.)

Lemma 2.7. *For functions $u_h, v_h \in \mathcal{H}_0^1(\mathcal{G}_h)$ we have*

$$
\big(\partial_k^+ u_h,\, v_h\big)_{\mathcal{L}^2(\mathcal{G}_h)} = -\big(u_h,\, \partial_k^- v_h\big)_{\mathcal{L}^2(\mathcal{G}_h)}, \quad k = 1, \dots, m,
$$

and

$$\left(u_h,\, u_h\right)_{\mathcal{L}^2(\mathcal{G}_h)} \leq C\left(\nabla^+ u_h,\, \nabla^+ u_h\right)_{(\mathcal{L}^2(\mathcal{G}_h))^m},$$

where the constant C *depends only on the domain* G*.*

Proof. The first assertion follows from a straightforward calculation. For the second one we refer to Temam (2001, Proposition 3.3 in Chapter 1). □

The dual space of $\mathcal{H}^1_0(\mathcal{G}_h)$ is denoted by $\mathcal{H}^{-1}(\mathcal{G}_h)$.

3. The results

3.1. Lipschitz-continuous reaction rates and linear diffusion.

3.1.1. *The macroscopic model.* In this section we describe a result for a classical system of reaction-diffusion equations, i.e., we assume, as in the general model, that there are n_r reactions going on, involving n_s species. Moreover, we assume that the diffusive mass fluxes $\boldsymbol{J}_j$ are given by Fick's law:

$$\boldsymbol{J}_j(\boldsymbol{u}, \nabla \boldsymbol{u}) = -D_j \nabla u_j, \quad j = 1, \ldots, n_s. \tag{8}$$

Here $D_1, \ldots, D_{n_s} > 0$ are the macroscopic diffusion coefficients. Hence, the macroscopic PDE system (with Dirichlet boundary conditions) reads

$$\begin{cases} \partial_t u_j - D_j \Delta u_j = f_j(\boldsymbol{u}) & \text{in } Q_T \\ u_j = 0 & \text{on } \partial G \times [0, T] \\ u_j(\cdot, 0) = u_{j,0} & \text{in } G, \end{cases} \tag{9}$$

$j = 1, \ldots, n_s$.

3.1.2. *The mesoscopic stochastic particle model.* A corresponding mesoscopic stochastic particle model is defined by setting $d_{j,k\pm} = d_j/(2m)$ with constant $d_j > 0$ in (5).

3.1.3. *Law of large numbers.* We make the following assumptions for the reaction rates K_i:

(10a) $K_i(\boldsymbol{v}) \geq 0$ for all $\boldsymbol{v} \in (\mathbb{R}^+_0)^{n_s}$.

(10b) If $\nu_{ij} < 0$ then $K_i(\boldsymbol{v}) = 0$ for all $\boldsymbol{v} \in (\mathbb{R}^+_0)^{n_s}$ with $v_j = 0$.

These two conditions should obviously be fulfilled by any set of reaction rates for physical reasons. In addition, the rates are supposed to satisfy the Lipschitz condition

$$|K_i(\boldsymbol{v}) - K_i(\boldsymbol{w})| \leq c_L\, |\boldsymbol{v} - \boldsymbol{w}|\,, \quad \boldsymbol{v}, \boldsymbol{w} \in \mathbb{R}^{n_s},\ i = 1, \ldots, n_r, \tag{10c}$$

for some constant $c_L > 0$, in order to ensure global existence and uniqueness of a solution.

We briefly describe the standard weak formulation of Eq. (9). We set

$$\boldsymbol{H}^1_0(G) = (H^1_0(G))^{n_s},\ \boldsymbol{L}^2(G) = (L^2(G))^{n_s},\ \boldsymbol{H}^{-1}(G) = ((H^1_0(G))^{n_s})^*,$$

and in the following we often skip the domain G in the notation. Let $a(\cdot,\cdot)$ be the bilinear form on $\boldsymbol{H}_0^1 \times \boldsymbol{H}_0^1$ defined by

$$a(\boldsymbol{u},\boldsymbol{v}) = \sum_{j=1}^{n_s}\sum_{k=1}^{m} D_j \big(\partial_{x_k} u_j,\, \partial_{x_k} v_j\big)_{L^2}, \quad \boldsymbol{u},\boldsymbol{v} \in \boldsymbol{H}_0^1. \tag{11}$$

In the weak formulation of Eq. (9), which is obtained by multiplying with a test function and integrating by parts, a function $\boldsymbol{u} \in H^1(0,T;\boldsymbol{H}_0^1,\boldsymbol{L}^2)$ is sought such that

$$\frac{d}{dt}\big(\boldsymbol{u}(t),\, \boldsymbol{v}\big)_{\boldsymbol{L}^2} + a(\boldsymbol{u}(t),\boldsymbol{v}) = \big(\boldsymbol{f}(\boldsymbol{u}),\, \boldsymbol{v}\big)_{\boldsymbol{L}^2} \tag{12a}$$

for all $\boldsymbol{v} \in \boldsymbol{H}_0^1$ and a.e. $t \in [0,T]$, and

$$\boldsymbol{u}(0) = \boldsymbol{u}_0 \in \boldsymbol{L}^2. \tag{12b}$$

Here $H^1(0,T;\boldsymbol{H}_0^1,\boldsymbol{L}^2)$ denotes the subspace of functions in $L^2(0,T;\boldsymbol{H}_0^1)$ that have generalised time derivatives in $L^2(0,T;\boldsymbol{H}^{-1})$, and Eq. (12a) is supposed to hold in the sense of distributions. The existence of a unique solution of the weak problem can readily be established with the Faedo-Galerkin method in combination with the Aubin-Lions compactness theorem (see, e.g., Lions (1969), Section 5 of Chapter 1). Alternatively, one can use the theory for linear equations together with the Banach fixed-point theorem (Evans, 1998).

For the passage to the limit, we assume the following scaling relations for the parameters in the stochastic particle model:

$$l \to 0, \; n \to \infty, \tag{13a}$$

$$\frac{d_j}{2m}\, l^2 \to D_j, \tag{13b}$$

$$\frac{d_j}{n} \to 0, \tag{13c}$$

$j = 1,\dots,n_s$. The law of large numbers then takes the following form.

Theorem 3.1 (Law of large numbers)**.** *Let $\boldsymbol{u}$ be the solution of the weak PDE problem* (12) *to the initial value $\boldsymbol{u}_0$. Assume that the scaling relations* (13) *are satisfied and that $\mathbf{u}_l(0)$ converges strongly to $\boldsymbol{u}_0$ in $\boldsymbol{L}^2$. Then*

$$E\left[\|\boldsymbol{u}_l - \boldsymbol{u}\|^2_{L^2(0,T;\boldsymbol{L}^2)}\right] = E\left[\|\boldsymbol{u}_l - \boldsymbol{u}\|^2_{(L^2(Q_T))^{n_s}}\right] \to 0.$$

3.2. **Crowding effects.** In this section we describe a result for the situation where the intensity for a diffusive jump of a particle increases with the density in the cell, i.e., the intensity for a jump to a neighboring cell is given by a function $d = d(u_l)$. The function d is assumed to be monotonously increasing, which models repulsive interactions between the particles. For the sake of simplicity we consider only a single-species model without chemical reactions.

3.2.1. *The macroscopic model.* The PDE that will be approached by the particle density process in the limit of large particle numbers is

$$\begin{cases} \partial_t u - \Delta(D(u)\,u) = 0 & \text{in } Q_T \\ u = 0 & \text{on } \partial G \times [0,T] \\ u(\cdot,0) = u_0 & \text{in } G, \end{cases} \tag{14}$$

where the function $D : \mathbb{R} \to \mathbb{R}_0^+$ is assumed to satisfy certain conditions that will be specified below. If we assume that the function D is differentiable, then Eq. (14) can be cast in the form (2) by setting

$$\boldsymbol{J}(u, \nabla u) = -\nabla\big(D(u)\,u\big) = -\big(uD'(u) + D(u)\big)\nabla u. \tag{15}$$

3.2.2. *The mesoscopic stochastic particle model.* A corresponding stochastic particle model is obtained by setting

$$d_{k-}(u_l, -\nabla^- u_l) = d_{k+}(u_l, \nabla^+ u_l) = d(u_l)/(2m), \quad k = 1, \dots, m, \tag{16}$$

in the general model for a monotonously increasing function $d : \mathbb{R} \to \mathbb{R}_0^+$. Further conditions on d will be specified below. Note that (for fixed l) by construction the process $u_l(t)$, $t \geq 0$, almost surely satisfies the two estimates

$$\sup_{z \in \mathcal{G}_l, t \geq 0} |u_l(z,t)| < \infty, \tag{17}$$

$$\big(u_l(t),\, 1\big)_{\mathcal{L}^2} = \|u_l(t)\|_{L^1(G)} \leq \|u_l(0)\|_{L^1(G)} \quad \text{for all } t \geq 0. \tag{18}$$

3.2.3. *Law of large numbers.* We start again by discussing an appropriate notion of weak solvability for Eq. (14) following Lions (1969, Section 3 of Chapter 2). Let the Hilbert space H_0^1 be endowed with the scalar product

$$\big(u,\, v\big)_{H_0^1} = \big(\nabla u,\, \nabla v\big)_{L^2}, \quad u, v \in H_0^1. \tag{19}$$

Hence, the operator $-\Delta : H_0^1 \to H^{-1}$, interpreted as

$$\big\langle -\Delta u,\, v\big\rangle_{H_0^1} = \big(\nabla u,\, \nabla v\big)_{(L^2)^m}, \quad u, v \in H_0^1, \tag{20}$$

is identical to the Riesz isomorphism between the Hilbert space H_0^1 and its dual H^{-1}. ($\langle \cdot,\, \cdot \rangle_{H_0^1}$ denotes the dual pairing between H_0^1 and H^{-1}.) Thus we can define on H^{-1} the scalar product

$$\big(u,\, v\big)_{H^{-1}} = \big\langle u,\, -\Delta^{-1} v\big\rangle_{H_0^1}, \quad u, v \in H^{-1}, \tag{21}$$

and we denote the corresponding norm by $|||\cdot|||_{H^{-1}}$. The norm $|||\cdot|||_{H^{-1}}$ is in fact equal to the standard norm in H^{-1} which is denoted by $\|\cdot\|_{H^{-1}}$. In order to ensure unique solvability of the weak problem introduced below, we make the following hypotheses for the function $D : \mathbb{R} \to \mathbb{R}_0^+$.

(22a) D is continuous and monotonously increasing on $\mathbb{R}_0^+$.

(22b) $D(p) = D(-p)$ for all $p \in \mathbb{R}$.

(22c) There are constants $C, \alpha > 0$ such that

$$D(p) \le C \text{ and } D(p)\,p^2 \ge \alpha\,p^2 \text{ for all } p \in \mathbb{R}.$$

It is then readily checked (Reichert, 2006) that the following lemma holds.

Lemma 3.2. *Let $a : L^2 \times L^2 \to \mathbb{R}$ be given by*

$$a(u,v) = \int_G D(u)\,u\,v\,dx, \quad u,v \in L^2,$$

and assume that D satisfies conditions (22)*. Then the mapping $a(\cdot,\cdot)$ induces a (generally nonlinear) operator $A : L^2 \to (L^2)^*$ by*

$$\big\langle A(u),\, v\big\rangle_{L^2} = a(u,v), \quad u,v \in L^2,$$

which is bounded, coercive, hemicontinuous and monotone.

For terminology see, e.g., Zeidler (1990). In the sequel, explicit use is made only of the monotonicity condition

$$\big\langle A(u) - A(v),\, u - v\big\rangle_{L^2} \ge 0, \quad u,v \in L^2. \tag{23}$$

In the weak formulation of the PDE (14) a function $u \in H^1(0,T;L^2,H^{-1})$ is sought such that

$$\frac{d}{dt}\big(u(t),\, v\big)_{H^{-1}} + a(u(t),v) = 0 \tag{24a}$$

for all $v \in L^2$ and a.e. $t \in [0,T]$, and

$$u(0) = u_0 \in H^{-1}. \tag{24b}$$

A unique weak solution exists according to a general theorem on monotone first-order evolution equations (see, e.g., Theorem 30.A in Zeidler (1990) or Theorem 1.2 in Chapter 2 of Lions (1969)).

For the derivation of a law of large numbers we suppose that the function $d : \mathbb{R} \to \mathbb{R}_0^+$ satisfies conditions (22a) and (22b). In addition, we make the following hypotheses:

$$l \to 0,\ n \to \infty, \tag{25a}$$

$$\sup_{\mathbb{R}} \big|\tfrac{1}{2m} l^2\, d - D\big| \to 0, \tag{25b}$$

$$\frac{1}{n} \sup_{\mathbb{R}} d \to 0. \tag{25c}$$

The law of large numbers then reads as follows.

Theorem 3.3 (Law of large numbers)**.** *Let u be the solution of the weak PDE problem* (24) *to the initial value $u_0 \in L^2$. Assume that the scaling relations* (25) *are satisfied and that $u_l(0)$ converges strongly to u_0 in L^2. Then the particle density u_l converges to u in the following sense: For all $\psi \in C_0^\infty(Q_T)$ and $\varepsilon > 0$,*

$$P\left[\left|\int_{Q_T} (u_l - u)\,\psi\,dx\,dt\right| > \varepsilon\right] \to 0.$$

With a little more effort it can be verified that the above theorem implies weak convergence of the random measures $u_l(x,t)\,dx\,dt$ on Q_T to the deterministic measure $u(x,t)\,dx\,dt$. (Weak convergence of random measures is discussed, e.g., in Daley & Vere-Jones (1988).)

3.3. **Gradient-activated diffusion.** In the present section we describe an example for nonlinear diffusion where the intensity for a diffusive jump to a neighboring cell increases with the concentration gradient. We again restrict the discussion to a single-species model without chemical reactions. The intensity for a jump in direction $\pm e_k$ is $d(\partial_k^+ u_l)$ and $d(-\partial_k^- u_l)$, respectively. It is again assumed that the function d satisfies $d(p) = d(-p)$ for $p \in \mathbb{R}$, i.e., the jump intensity changes according to the absolute value of the concentration gradient. We call this behaviour gradient-activated diffusion.

3.3.1. *The macroscopic model.* The macroscopic PDE that will be approached in the limit of large particle numbers reads

$$\begin{cases} \partial_t u - \sum_{k=1}^{m} \partial_{x_k}(D(\partial_{x_k} u)\,\partial_{x_k} u) = 0 & \text{in } Q_T \\ u = 0 & \text{on } \partial G \times [0,T] \\ u(0) = u_0 & \text{in } G. \end{cases} \tag{26}$$

We assume again that D satisfies conditions (22). The PDE (26) can be cast in the form (2) by setting

$$J_k(u, \nabla u) = -D(\partial_{x_k})\,\partial_{x_k} u, \quad k = 1, \ldots, m. \tag{27}$$

3.3.2. *The mesoscopic stochastic particle model.* A corresponding stochastic particle model is given by setting

$$d_{k-}(u_l, -\nabla^- u_l) = d(-\partial_k^- u_l), \tag{28}$$

$$d_{k+}(u_l, \nabla^+ u_l) = d(\partial_k^+ u_l), \tag{29}$$

for a function $d : \mathbb{R} \to \mathbb{R}_0^+$ that satisfies (22a) and (22b).

3.3.3. *Law of large numbers.* In order to discuss the existence of a solution, we return here to the functional setting of Section (3.1), i.e., we look for a function in $H^1(0,T; H_0^1, L^2)$ that solves an appropriate weak formulation of Eq. (26). Again a monotonicity property plays a crucial role.

Lemma 3.4. *Let $a : H_0^1 \times H_0^1 \to \mathbb{R}$ be given by*

$$a(u,v) = \sum_{k=1}^{m} \int_G D(\partial_{x_k} u)\,\partial_{x_k} u\,\partial_{x_k} v\,dx, \quad u, v \in H_0^1,$$

and assume that D satisfies conditions (22) *of the previous section. Then the mapping $a(\cdot,\cdot)$ induces a (generally nonlinear) operator $A : H_0^1 \to H^{-1}$ by*

$$\langle A(u),\, v \rangle_{H_0^1} = a(u,v), \quad u, v \in H_0^1,$$

which is bounded, coercive, hemicontinuous and monotone.

Proof. The proof is similar to the proof of Lemma 3.2. □

The weak formulation of the PDE is obtained in the usual way by multiplying Eq. (14) with a test function and integrating by parts:

$$\frac{d}{dt}\big(u(t),\, v\big)_{L^2} + a(u(t), v) = 0 \tag{30a}$$

for all $v \in H_0^1$ and a.e. $t \in [0, T]$, and

$$u(0) = u_0 \in L^2. \tag{30b}$$

Again a general theorem on first-order monotone evolution equations (Zeidler, 1990; Lions, 1969) ensures that the weak problem (30) has a unique solution.

We assume the following scaling relations:

$$l \to 0,\; n \to \infty, \tag{31a}$$

$$\sup_{\mathbb{R}} \left| l^2\, d - D \right| \to 0, \tag{31b}$$

$$\frac{1}{n} \sup_{\mathbb{R}} d \to 0. \tag{31c}$$

The law of large numbers then reads as follows.

Theorem 3.5 (Law of large numbers)**.** *Let u be the solution of the weak PDE problem* (30) *to the initial value u_0. Assume that the scaling relations* (31) *are satisfied and that $u_l(0)$ converges strongly to u_0 in L^2. Then*

$$E\left[\|u_l - u\|^2_{L^2(0,T;L^2)}\right] = E\left[\|u_l - u\|^2_{L^2(Q_T)}\right] \to 0.$$

4. The convergence proofs

To prove convergence of the stochastic particle densities we consider first an auxiliary problem where the spatial derivatives in the continuum model are replaced by finite differences. In a second step we show that the difference between the stochastic process and the solution of the auxiliary problem converges to zero.

4.1. **Lipschitz continuous reaction rates and linear diffusion.**

4.1.1. *An auxiliary problem.* Let $\mathcal{G}_h$ be the interior lattice points generated by a lattice $\mathcal{Z}_h(z_0)$ of the domain G representing the chemical reactor, and let

$$\boldsymbol{\mathcal{L}}^2(\mathcal{G}_h) = (\mathcal{L}^2(\mathcal{G}_h))^{n_s},\; \boldsymbol{\mathcal{H}}_0^1(\mathcal{G}_h) = (\mathcal{H}_0^1(\mathcal{G}_h))^{n_s},\; \boldsymbol{\mathcal{H}}^{-1}(\mathcal{G}_h) = (\boldsymbol{\mathcal{H}}_0^1(\mathcal{G}_h))^*,$$

be the discrete versions of $\boldsymbol{L}^2(G)$, $\boldsymbol{H}_0^1(G)$, and $\boldsymbol{H}^{-1}(G)$. A discrete analogue of the PDE system (9) is given by

$$\begin{cases} u_{h,j}' - D_{h,j}\nabla^- \cdot \nabla^+ u_{h,j} = f_j(\boldsymbol{u}_h) & \text{in } \mathcal{G}_h^1 \times (0,T) \\ u_{h,j} = 0 & \text{on } (\mathcal{G}_h \setminus \mathcal{G}_h^1) \times [0,T] \\ u_{h,j} = u_{h,j,0} & \text{in } \mathcal{G}_h^1, \end{cases} \tag{32}$$

$j = 1, \ldots, n_s$, with constants $D_{h,j} > 0$, which is an initial-value problem for a (nonlinear) finite-dimensional system of ODEs with (globally) Lipschitz continuous right-hand side. Hence, according to the Picard-Lindelöf theorem, it has a unique solution on the entire interval $[0, T]$.

The discrete version $a_h(\cdot, \cdot)$ of the bilinear form $a(\cdot, \cdot)$ defined in (11) is

$$a_h(\boldsymbol{u}_h, \boldsymbol{v}_h) = \sum_{j=1}^{n_s} \sum_{k=1}^{m} D_{h,j} \big(\partial_k^+ u_{h,j},\, \partial_k^+ v_{h,j}\big)_{\mathcal{L}^2}, \quad \boldsymbol{u}_h, \boldsymbol{v}_h \in \boldsymbol{\mathcal{H}}_0^1, \tag{33}$$

and the solution of (32) can be regarded as a function in $C^1([0, T], \boldsymbol{\mathcal{H}}_0^1)$ that satisfies the following discrete version of the weak formulation (12):

$$\frac{d}{dt}\big(\boldsymbol{u}_h(t),\, \boldsymbol{v}_h\big)_{\boldsymbol{\mathcal{L}}^2} + a_h(\boldsymbol{u}_h(t), \boldsymbol{v}_h) = \big(\boldsymbol{f}(\boldsymbol{u}_h(t)),\, \boldsymbol{v}_h\big)_{\boldsymbol{\mathcal{L}}^2} \tag{34}$$

for all $\boldsymbol{v}_h \in \boldsymbol{\mathcal{H}}_0^1$ and $t \in [0, T]$. Note that the bilinear form $a_h(\cdot, \cdot)$ is coercive, i.e.,

$$a_h(\boldsymbol{u}_h, \boldsymbol{u}_h) \geq \alpha \|\boldsymbol{u}_h\|^2_{\boldsymbol{\mathcal{H}}_0^1} \tag{35}$$

for a constant $\alpha > 0$ because of the discrete Poincaré inequality.

The solution of the auxiliary problem (32) converges to the solution of the weak problem (12) in the following sense.

Theorem 4.1. *Let $\boldsymbol{u}$ be the solution of the weak PDE problem* (12) *to the initial value $\boldsymbol{u}_0$, and let $(\boldsymbol{u}_h)$, $h \searrow 0$, be a sequence of solutions of the approximating problem* (32) *to the initial value $\boldsymbol{u}_{h,0}$. Assume $\boldsymbol{u}_{h,0} \to \boldsymbol{u}_0$ strongly in $\boldsymbol{L}^2$, and $D_{h,j} \to D_j$, $j = 1, \ldots, n_s$. Then $\boldsymbol{u}_h$ converges strongly to $\boldsymbol{u}$ in $L^2(0, T; \boldsymbol{L}^2)$.*

Sketch of proof. The proof can be carried out in analogy to the existence proof for the weak problem (12) with the Faedo-Galerkin method by making use of the methodology of 'external approximations' which is introduced, e.g., in Temam (2001, Chapter 1) or Zeidler (1990, Chapter 35). Since the details of the calculations are rather uninteresting, we give only a short sketch. More information can be found in Reichert (2006).

By inserting $\boldsymbol{u}_h(t)$ for $\boldsymbol{v}_h$ in the discrete weak formulation, integrating over time and making use of the coerciveness of the bilinear form $a_h(\cdot, \cdot)$, we get

$$\|\boldsymbol{u}_h(t)\|^2_{\boldsymbol{\mathcal{L}}^2} + 2\alpha \int_0^t \|\boldsymbol{u}_h(s)\|^2_{\boldsymbol{\mathcal{H}}_0^1}\, ds \leq \|\boldsymbol{u}_h(0)\|^2_{\boldsymbol{\mathcal{L}}^2} + \\ + 2 \int_0^t \big(\boldsymbol{f}(\boldsymbol{u}_h(s)),\, \boldsymbol{u}_h(s)\big)_{\boldsymbol{\mathcal{L}}^2}\, ds. \tag{36}$$

Hence, in view of the Lipschitz condition (10c), we have the a-priori estimates

$$\sup_h \max_{0 \leq t \leq T} \|\boldsymbol{u}_h(t)\|_{\boldsymbol{\mathcal{L}}^2} < \infty, \tag{37}$$

$$\sup_h \|\boldsymbol{u}_h\|_{L^2(0,T;\boldsymbol{\mathcal{H}}_0^1)} < \infty, \tag{38}$$

$$\sup_h \|\boldsymbol{u}_h'\|_{L^2(0,T;\boldsymbol{\mathcal{H}}^{-1})} < \infty. \tag{39}$$

The weak convergence in $L^2(0,T;\boldsymbol{L}^2)$ of the sequence $(\boldsymbol{u}_h)$ to the solution of (12) can now be established with techniques from Temam (2001). The strong convergence follows from a discrete analogue of the Aubin-Lions compactness theorem. □

4.1.2. *Convergence of the particle density.* Henceforth we denote the stochastic particle density by $\boldsymbol{u}_l$ and the solutions of the auxiliary approximating problem (32) with $h = l$ by $\boldsymbol{v}_l$. In view of Theorem 4.1, the law of large numbers (Theorem 3.1) follows immediately from the next result.

Theorem 4.2. *Let $\boldsymbol{v}_l$ be the solutions of the auxiliary approximating problem (32) to the initial value $\boldsymbol{v}_{l,0}$ with $D_{l,j} = \frac{d_j}{2m} l^2$, $j = 1, \dots, n_s$. Moreover, assume that $\|\boldsymbol{u}_l(0) - \boldsymbol{v}_{l,0}\|_{\boldsymbol{\mathcal{L}}^2} \to 0$. Then*

$$\sup_{t \le T} E\Big[\|\boldsymbol{u}_l(t) - \boldsymbol{v}_l(t)\|_{\boldsymbol{\mathcal{L}}^2}^2\Big] \to 0.$$

The proof of Theorem 4.2 is based on a lemma that identifies a local martingale associated to the process $\|\boldsymbol{u}_l(t) - \boldsymbol{v}_l(t)\|_{\boldsymbol{\mathcal{L}}^2}^2$, $t \le T$. Before stating and proving this lemma we have to deal with the slight technical difficulty that reactive jumps that would lead out of the state space, i.e., to negative particle densities, are not 'automatically' excluded. It might happen that a $K_i(\boldsymbol{w})$ is positive for a certain vector of densities $\boldsymbol{w} \in \frac{1}{n}\mathbb{N}_0^{n_s}$ although the transition from a state $\boldsymbol{u}_l \in S_l$ with $\boldsymbol{u}_l(z) = \boldsymbol{w}$ for a $z \in \mathcal{G}_l$ to $\tilde{\boldsymbol{u}}_l = \boldsymbol{u}_l + \frac{1}{n}\sum_{j=1}^{n_s} \nu_{ij} \boldsymbol{\chi}_{j,z}$ (i.e., $\tilde{u}_{l,j}(z) = w_j + \frac{1}{n}\nu_{ij}$ for $j = 1, \dots, n_s$) is not allowed because it would lead to negative particle densities. This might be the case if w_j is close to zero for a certain j and ν_{ij} is negative, say, $w_j = 1/n$ and $\nu_{ij} = -2$. However, we may always assume (by possibly modifying the original K_i) that there are measurable functions $K_{l,i} : \mathbb{R}^{n_s} \to \mathbb{R}$, $i = 1, \dots, n_r$, that converge uniformly to K_i for $l \to 0$, such that the transition intensities are left unchanged and intensity zero is automatically assigned to jumps that would leave the state space:

$$K_{l,i}(\boldsymbol{w}) = \begin{cases} K_i(\boldsymbol{w}) & \text{if} \quad w_j + \frac{1}{n}\nu_{ij} \ge 0 \text{ for all } j = 1, \dots, n_s \\ 0 & \text{otherwise} \end{cases} \tag{40}$$

for all $\boldsymbol{w} \in \frac{1}{n}\mathbb{N}_0^{n_s}$, and

$$\sup_{R^{n_s}} |K_i - K_{l,i}| \to 0 \quad (l \to 0). \tag{41}$$

The vector of reaction functions corresponding to the modified rates $K_{l,i}$, which is defined in the same way as in Eq. (3), is denoted by $\boldsymbol{f}_l$.

We define for $p \in \mathbb{N}$ the stopping time τ_p by

$$\tau_p = \inf \Big\{ t : \|\boldsymbol{u}_l(t)\|_{\boldsymbol{\mathcal{L}}^2} > p \Big\} \wedge T. \tag{42}$$

We then have the following lemma.

Lemma 4.3. *Let* $(M_l(t))_{t\le T}$ *be the process given by*

$$\begin{aligned} M_l(t) = &\|\boldsymbol{u}_l(t)-\boldsymbol{v}_l(t)\|^2_{\boldsymbol{\mathcal{L}}^2} - \|\boldsymbol{u}_l(0)-\boldsymbol{v}_l(0)\|^2_{\boldsymbol{\mathcal{L}}^2}+\\ &+2\int_0^t a_l(\boldsymbol{u}_l(s)-\boldsymbol{v}_l(s),\boldsymbol{u}_l(s)-\boldsymbol{v}_l(s))\,ds-\\ &-2\int_0^t \Big(\boldsymbol{f}_l(\boldsymbol{u}_l(s))-\boldsymbol{f}(\boldsymbol{v}_l(s)),\,\boldsymbol{u}_l(s)-\boldsymbol{v}_l(s)\Big)_{\boldsymbol{\mathcal{L}}^2}\,ds - R_l(t), \end{aligned} \tag{43}$$

where

$$\begin{aligned} R_l(t) = &2\sum_{j=1}^{n_s}\frac{d_j}{n}\int_0^t \big(u_{l,j}(s),\,1\big)_{\mathcal{L}^2}\,ds+\\ &+\frac{1}{n}\sum_{i=1}^{n_r}\sum_{j=1}^{n_s}\nu_{ij}^2\int_0^t\big(K_{l,i}(\boldsymbol{u}_l(s)),\,1\big)_{\mathcal{L}^2}\,ds. \end{aligned} \tag{44}$$

Then, for each $p\in\mathbb{N}$*, the stopped process* $M_l(t\wedge\tau_p))_{t\le T}$ *is a martingale.*

Proof. 1. For arbitrary but fixed $\boldsymbol{w}_l\in\boldsymbol{\mathcal{H}}_0^1$ we define the function $g(\cdot,\boldsymbol{w}_l): S_l\to\mathbb{R}$, $\boldsymbol{u}_l\mapsto g(\boldsymbol{u}_l,\boldsymbol{w}_l)=\|\boldsymbol{u}_l-\boldsymbol{w}_l\|^2_{\boldsymbol{\mathcal{L}}^2}$, and we are going to compute $L_lg(\boldsymbol{u}_l,\boldsymbol{w}_l)$. The generator L_l can be written as $L_l=L_{d,l}+L_{r,l}$ if we separate jumps due to reaction and diffusion events in the sum (7). We start with the computation of $L_{d,l}g(\boldsymbol{u}_l,\boldsymbol{w}_l)$.

$$\begin{aligned} L_{d,l}g(\boldsymbol{u}_l,\boldsymbol{w}_l) &= \sum_{j=1}^{n_s}\sum_{z\in\mathcal{G}_l^1}\sum_{k=1}^{m} n\,\frac{d_j}{2m}\,u_{l,j}(z)\times\\ &\quad\times\Big(\|\boldsymbol{u}_l-\tfrac{1}{n}\boldsymbol{\chi}_{j,z}+\tfrac{1}{n}\boldsymbol{\chi}_{j,(z-le_k)}-\boldsymbol{w}_l\|^2_{\boldsymbol{\mathcal{L}}^2}-\\ &\quad-2\,\|\boldsymbol{u}_l-\boldsymbol{w}_l\|^2_{\boldsymbol{\mathcal{L}}^2}+\\ &\quad+\|\boldsymbol{u}_l-\tfrac{1}{n}\boldsymbol{\chi}_{j,z}+\tfrac{1}{n}\boldsymbol{\chi}_{j,(z+le_k)}-\boldsymbol{w}_l\|^2_{\boldsymbol{\mathcal{L}}^2}\Big)\\ &=\sum_{j=1}^{n_s}\sum_{z\in\mathcal{G}_l^1}\sum_{k=1}^{m} n\,\frac{d_j}{2m}\,u_{l,j}(z)\times\\ &\quad\times\bigg(2\frac{l^{m+2}}{n}\Big(\partial_k^-\partial_k^+u_{l,j}(z)-\partial_k^-\partial_k^+w_{l,j}(z)\Big)+4\frac{l^m}{n^2}\bigg)\\ &=\sum_{j=1}^{n_s}\sum_{k=1}^{m}\bigg(2\,\frac{d_j}{2m}\,l^2\big(u_{l,j},\,\partial_k^-\partial_k^+\,(u_{l,j}-w_{l,j})\big)_{\mathcal{L}^2}+\\ &\quad+\frac{2}{m}\,\frac{d_j}{n}\,\big(u_{l,j},\,1\big)_{\mathcal{L}^2}\bigg)\\ &=-2\sum_{j=1}^{n_s}\sum_{k=1}^{m}\frac{d_j}{2m}\,l^2\,\big(\partial_k^+u_{l,j},\,\partial_k^+(u_{l,j}-w_{l,j})\big)_{\mathcal{L}^2}+ \end{aligned} \tag{45}$$

$$+\,2\sum_{j=1}^{n_s}\frac{d_j}{n}\big(u_{l,j},\,1\big)_{\mathcal{L}^2}$$
$$=-2\,a_l(\boldsymbol{u}_l,\boldsymbol{u}_l-\boldsymbol{w}_l)+2\sum_{j=1}^{n_s}\frac{d_j}{n}\big(u_{l,j},\,1\big)_{\mathcal{L}^2}.$$

Here $a_l(\cdot,\cdot)$ is given by (33) with $D_{l,j}=\frac{d_j}{2m}\,l^2$. Computing the reaction part yields

$$\begin{aligned}L_{r,l}g(\boldsymbol{u}_l,\boldsymbol{w}_l)&=\sum_{z\in\mathcal{G}_l^1}\sum_{i=1}^{n_r}n\,K_{l,i}(\boldsymbol{u}_l(z))\times\\&\quad\times\Big(\|\boldsymbol{u}_l+\tfrac{1}{n}\textstyle\sum_{j=1}^{n_s}\nu_{ij}\boldsymbol{\chi}_{j,z}-\boldsymbol{w}_l\|^2_{\boldsymbol{\mathcal{L}}^2}-\\&\qquad-\|\boldsymbol{u}_l-\boldsymbol{w}_l\|^2_{\boldsymbol{\mathcal{L}}^2}\Big)\\&=\sum_{z\in\mathcal{G}_l^1}\sum_{i=1}^{n_r}n\,K_{l,i}(\boldsymbol{u}_l(z))\times\\&\quad\times\left(2\frac{l^m}{n}\sum_{j=1}^{n_s}\nu_{ij}\,(u_{l,j}(z)-w_{l,j}(z))+\frac{l^m}{n^2}\sum_{j=1}^{n_s}\nu_{ij}^2\right).\end{aligned}\tag{46}$$

Hence,

$$\begin{aligned}L_{r,l}g(\boldsymbol{u}_l,\boldsymbol{w}_l)&=\sum_{i=1}^{n_r}\sum_{j=1}^{n_s}\sum_{z\in\mathcal{G}_l^1}\bigg(2\,n\frac{l^m}{n}\,\nu_{ij}\,K_{l,i}(\boldsymbol{u}_l(z))\big(u_{l,j}(z)-w_{l,j}(z)\big)+\\&\qquad+n\frac{l^m}{n^2}\,\nu_{ij}^2\,K_{l,i}(\boldsymbol{u}_l(z))\bigg)\\&=2\left(\boldsymbol{f}_l(\boldsymbol{u}_l),\,\boldsymbol{u}_l-\boldsymbol{w}_l\right)_{\boldsymbol{\mathcal{L}}^2}+\frac{1}{n}\sum_{i=1}^{n_r}\sum_{j=1}^{n_s}\nu_{ij}^2\left(K_{l,i}(\boldsymbol{u}_l),\,1\right)_{\mathcal{L}^2}.\end{aligned}\tag{47}$$

By gathering together the different contributions we finally get

$$\begin{aligned}L_lg(\boldsymbol{u}_l,\boldsymbol{w}_l)&=-2\,a_l(\boldsymbol{u}_l,\boldsymbol{u}_l-\boldsymbol{w}_l)+2\left(\boldsymbol{f}_l(\boldsymbol{u}_l),\,\boldsymbol{u}_l-\boldsymbol{w}_l\right)_{\boldsymbol{\mathcal{L}}^2}+\\&\quad+2\sum_{j=1}^{n_s}\frac{d_j}{n}\big(u_{l,j},\,1\big)_{\mathcal{L}^2}+\frac{1}{n}\sum_{i=1}^{n_r}\sum_{j=1}^{n_s}\nu_{ij}^2\left(K_{l,i}(\boldsymbol{u}_l),\,1\right)_{\mathcal{L}^2}.\end{aligned}\tag{48}$$

Recall that we denote by $\boldsymbol{v}_l(t)$ the solutions of the approximating problem (32), and consider the function $h(\cdot,\boldsymbol{w}_l):[0,T]\to\mathbb{R}$ with $\boldsymbol{w}_l\in\boldsymbol{\mathcal{H}}_0^1$ as parameter given by $t\mapsto h(t,\boldsymbol{w}_l)=\|\boldsymbol{w}_l-\boldsymbol{v}_l(t)\|^2_{\boldsymbol{\mathcal{L}}^2}$. Note that

$$\begin{aligned}h'(t,\boldsymbol{w}_l)&=-2\left(\boldsymbol{v}_l'(t),\,\boldsymbol{w}_l-\boldsymbol{v}_l(t)\right)_{\boldsymbol{\mathcal{L}}^2}\\&=2\,a_l\big(\boldsymbol{v}_l(t),\boldsymbol{w}_l-\boldsymbol{v}_l(t)\big)-2\left(\boldsymbol{f}(\boldsymbol{v}_l(t)),\,\boldsymbol{w}_l-\boldsymbol{v}_l(t)\right)_{\boldsymbol{\mathcal{L}}^2}.\end{aligned}$$

2. Consider now the (unbounded) function $\Phi : S_l \times [0,T] \to \mathbb{R}$ given by

$$(\boldsymbol{u}_l, t) \mapsto \Phi(\boldsymbol{u}_l, t) = \|\boldsymbol{u}_l - \boldsymbol{v}_l(t)\|^2_{\boldsymbol{\mathcal{L}}^2}. \tag{49}$$

It follows by Dynkin's formula (Kallenberg, 2002, Lemma 19.21) and a truncation argument that the process

$$\begin{aligned} M_l(t) &= \|\boldsymbol{u}_l(t) - \boldsymbol{v}_l(t)\|^2_{\boldsymbol{\mathcal{L}}^2} - \|\boldsymbol{u}_l(0) - \boldsymbol{v}_l(0)\|^2_{\boldsymbol{\mathcal{L}}^2} - \\ &\quad - \int_0^t \big(L_l\Phi(\boldsymbol{u}_l(s), s) + \partial_s\Phi(\boldsymbol{u}_l(s), s)\big)\, ds \\ &= \|\boldsymbol{u}_l(t) - \boldsymbol{v}_l(t)\|^2_{\boldsymbol{\mathcal{L}}^2} - \|\boldsymbol{u}_l(0) - \boldsymbol{v}_l(0)\|^2_{\boldsymbol{\mathcal{L}}^2} - \\ &\quad - \int_0^t \big(L_l g(\boldsymbol{u}_l(s), \boldsymbol{v}_l(s)) + h'(s, \boldsymbol{u}_l(s))\big)\, ds, \end{aligned} \tag{50}$$

$t \le T$, stopped at τ_p, is a martingale for each $p \in \mathbb{N}$. Plugging the explicit computations above into Eq. (50) yields (43). □

We are now ready to finish the proof of the law of large numbers.

Proof of Theorem 3.1. Let $\hat{d} = \max_{j=1,\dots,n_s} d_j$. By stopping the local martingale (43) and taking expectations we get the estimate

$$\begin{aligned} &E\Big[\|\boldsymbol{u}_l(t\wedge\tau_p) - \boldsymbol{v}_l(t\wedge\tau_p)\|^2_{\boldsymbol{\mathcal{L}}^2}\Big] + \\ &\qquad + E\int_0^{t\wedge\tau_p} a_l\big(\boldsymbol{u}_l(s) - \boldsymbol{v}_l(s), \boldsymbol{u}_l(s) - \boldsymbol{v}_l(s)\big)\, ds \\ &\quad \le \|\boldsymbol{u}_l(0) - \boldsymbol{v}_l(0)\|^2_{\boldsymbol{\mathcal{L}}^2} + \\ &\qquad + E\int_0^{t\wedge\tau_p} \Big|\big(\boldsymbol{f}_l(\boldsymbol{u}_l(s)) - \boldsymbol{f}(\boldsymbol{v}_l(s)),\, \boldsymbol{u}_l(s) - \boldsymbol{v}_l(s)\big)_{\boldsymbol{\mathcal{L}}^2}\Big|\, ds + \\ &\qquad + 2\sum_{j=1}^{n_s} \frac{d_j}{n} E\int_0^{t\wedge\tau_p} \big(u_{l,j}(s),\, 1\big)_{\mathcal{L}^2}\, ds + \\ &\qquad + \frac{1}{n}\sum_{j=1}^{n_s}\sum_{i=1}^{n_r} \nu_{ij}^2\, E\int_0^{t\wedge\tau_p} \big(K_i(\boldsymbol{u}_l(s)),\, 1\big)_{\mathcal{L}^2}\, ds. \end{aligned} \tag{51}$$

From the coerciveness of the bilinear form $a_l(\cdot,\cdot)$ and a few elementary estimates it follows that

$$\begin{aligned} &E\Big[\|\boldsymbol{u}_l(t\wedge\tau_p) - \boldsymbol{v}_l(t\wedge\tau_p)\|^2_{\boldsymbol{\mathcal{L}}^2}\Big] \\ &\quad \le \|\boldsymbol{u}_l(0) - \boldsymbol{v}_l(0)\|^2_{\boldsymbol{\mathcal{L}}^2} + C\sup_{\mathbb{R}^{n_S}} |\boldsymbol{f}_l - \boldsymbol{f}| + \\ &\qquad + C\,\big((\hat{d} + C\,)/n\big) \int_0^T \Big(\|\boldsymbol{v}_l(s)\|^2_{\boldsymbol{\mathcal{L}}^2} + 1\Big)\, ds + \\ &\qquad + C\,\Big((\hat{d} + C\,)/n + \sup_{\mathbb{R}^{n_S}} |\boldsymbol{f}_l - \boldsymbol{f}| + 1\Big) \times \\ &\qquad \times E\int_0^{t\wedge\tau_p} \|\boldsymbol{u}_l(s) - \boldsymbol{v}_l(s)\|^2_{\boldsymbol{\mathcal{L}}^2}\, ds, \end{aligned} \tag{52}$$

where the constant C does not depend on l. Note also that

$$\int_0^T E\Big[\|\boldsymbol{u}_l(s)\|^2_{\boldsymbol{\mathcal{L}}^2}\Big] < \infty. \tag{53}$$

By letting $p \to \infty$ we deduce from the monotone convergence theorem and Fatou's lemma that the above estimate (52) is valid even with $t \wedge \tau_p$ replaced by t. Gronwall's inequality then yields

$$\begin{aligned} &E\Big[\|\boldsymbol{u}_l(t) - \boldsymbol{v}_l(t)\|^2_{\boldsymbol{\mathcal{L}}^2}\Big] \\ &\quad \le \bigg(\|\boldsymbol{u}_l(0) - \boldsymbol{v}_l(0)\|^2_{\mathcal{L}^2} + C \sup_{\mathbb{R}^n S} |\boldsymbol{f}_l - \boldsymbol{f}| + \\ &\qquad + C\,\big((\hat{d} + C\,)/n\big)\Big(\|v_l(s)\|^2_{L^2(0,T;\boldsymbol{\mathcal{L}}^2)} + T\Big)\bigg) \times \\ &\qquad \times \exp\Big(C\,\big((\hat{d} + C\,)/n + \sup_{\mathbb{R}^n S} |\boldsymbol{f}_l - \boldsymbol{f}| + 1\big)\,T\Big). \end{aligned} \tag{54}$$

Finally, it follows from the scaling assumptions (13) and (41) that

$$\sup_{t \le T} E\Big[\|\boldsymbol{u}_l(t) - \boldsymbol{v}_l(t)\|^2_{\boldsymbol{\mathcal{L}}^2}\Big] \to 0. \tag{55}$$

□

4.2. **Crowding effects.**

4.2.1. *An auxiliary problem.* Here the discrete analogue of the PDE (14) on the interior lattice points $\mathcal{G}_h$ is given by

$$\begin{cases} u_h' - \Delta_h(D_h(u_h)\,u_h) = 0 & \text{in } \mathcal{G}_h^1 \times (0,T) \\ u_h = 0 & \text{on } (\mathcal{G}_h \setminus \mathcal{G}_h^1) \times [0,T] \\ u_h(\cdot, 0) = u_{h,0} & \text{on } \mathcal{G}_h^1, \end{cases} \tag{56}$$

where $\Delta_h = \nabla^- \cdot \nabla^+$ is the discrete Laplacian. We assume that the functions D_h satisfy conditions (22a) and (22b) and that

$$\sup_{\mathbb{R}} |D_h - D| \to 0 \quad (h \to 0). \tag{57}$$

The discretised PDE (56) is in fact a system of ODEs with continuous right-hand side which has a local solution according to the Peano theorem. Existence of a solution on the whole interval $[0,T]$ follows from the derivation of the a-priori estimate (64) below.

In analogy to the treatment of the PDE we endow the discrete Sobolev space $\mathcal{H}_0^1$ with the scalar product

$$\big(u_h,\, v_h\big)_{\mathcal{H}_0^1} = \big(\nabla^+ u_h,\, \nabla^+ v_h\big)_{(\mathcal{L}^2)^m}, \quad u_h, v_h \in \mathcal{H}_0^1. \tag{58}$$

It induces a norm which is equivalent to the original one due to the discrete Poincaré inequality. We regard $-\Delta_h$ as operator from $\mathcal{H}_0^1$ to $\mathcal{H}^{-1}$ given by

$$\big\langle -\Delta_h u_h,\, v_h\big\rangle_{\mathcal{H}_0^1} = \big(\nabla^+ u_h,\, \nabla^+ v_h\big)_{(\mathcal{L}^2)^m}, \quad u_h, v_h \in \mathcal{H}_0^1, \tag{59}$$

and $\mathcal{H}^{-1}$ is equipped with the scalar product

$$(u_h, v_h)_{\mathcal{H}^{-1}} = \langle u_h, -\Delta_h^{-1} v_h \rangle_{\mathcal{H}_0^1}, \quad u_h, v_h \in \mathcal{H}^{-1}. \tag{60}$$

The corresponding norm is denoted by $|||\cdot|||_{\mathcal{H}^{-1}}$. It is equal to the standard norm which we denote by $\|\cdot\|_{\mathcal{H}^{-1}}$. The solution of (56) can then be regarded as a function in $C^1([0,T],\mathcal{L}^2)$ that satisfies

$$\frac{d}{dt}(u_h(t), v_h)_{\mathcal{H}^{-1}} + a_h(u_h(t), v_h) = 0 \tag{61}$$

for all $v_h \in \mathcal{L}^2$ and $t \in [0,T]$, where the mapping $a_h : \mathcal{L}^2 \times \mathcal{L}^2 \to \mathbb{R}$ is defined by

$$a_h(u_h, v_h) = (D_h(u_h)\, u_h, v_h)_{\mathcal{L}^2}, \quad u_h, v_h \in \mathcal{L}^2. \tag{62}$$

Note that $a_h(\cdot,\cdot)$ induces a bounded monotone operator $A_h : \mathcal{L}^2 \to (\mathcal{L}^2)^*$ by

$$\langle A_h(u_h), v_h \rangle_{\mathcal{L}^2} = a_h(u_h, v_h), \quad u_h, v_h \in \mathcal{L}^2. \tag{63}$$

Compared to the example of the previous section we have introduced here a different functional setting to establish the existence of a unique weak solution of the macroscopic PDE (cf. Section. 3.2.3). Unfortunately, we are (without further hypotheses and regularity considerations) only able to show weak convergence of the solution of the auxiliary problem (56) in the space $L^2(0,T;L^2)$, which, in turn, results in a weaker law of large numbers.

Theorem 4.4. *Let u be the solution of the weak problem* (24) *to the initial value $u_0 \in L^2$. Let (u_h), $h \searrow 0$, be a sequence of solutions of the approximating problem* (61) *to the initial value $u_{h,0}$, and assume that $u_{h,0}$ converges strongly to u_0 in L^2. Then u_h converges weakly to u in $L^2(0,T;L^2)$.*

Sketch of proof. The a-priori estimates

$$\sup_h \max_{0\le t\le T} \|u_h(t)\|_{\mathcal{H}^{-1}} < \infty, \tag{64}$$

$$\sup_h \|u_h\|_{L^2(0,T;\mathcal{L}^2)} < \infty, \tag{65}$$

follow immediately from the discrete weak formulation by inserting $u_h(t)$ for v_h and integrating over time. They ensure the existence of a weakly convergent subsequence of (u_h) in $L^2(0,T;L^2)$. The passage to the limit can be performed with the aid of techniques from (Temam, 2001) and the Minty lemma. For more details see Reichert (2006). □

4.2.2. *Convergence of the particle density.* Let from now on v_l be the solution of the approximating problem (56) and u_l the stochastic particle density. We shall see below that by similar arguments as in the previous section we are able to show that

$$\sup_{t\le T} E\Big[\|u_l(t) - v_l(t)\|^2_{\mathcal{H}^{-1}}\Big] \to 0 \quad (l \to 0). \tag{66}$$

Unfortunately, we do not have a nice compatibility of the norms in $\mathcal{H}^{-1}$ and H^{-1}. If a sequence of lattice functions (u_h) converges to zero with respect to the norm in $\mathcal{H}^{-1}(\mathcal{G}_h)$ we are (to the best of our knowledge) not able to conclude that the same is true for the extended versions $(\tilde{u}_h|_G)$ with respect to the norm in $H^{-1}(G)$. This difficulty is circumvented by resorting to a weaker notion of convergence.

Observe that if we define for a function $\psi \in C_0^\infty(Q_T)$ approximating lattice functions $\psi_h(\cdot, t)$ on a grid $\mathcal{Z}_h(z_0)$ simply by setting

$$\psi_h(z,t) = \begin{cases} \psi(z,t) & \text{for } z \in \mathcal{G}_h^1 \\ 0 & \text{otherwise,} \end{cases} \tag{67}$$

then $\tilde{\psi}_h|_{Q_T}$ converges uniformly to ψ on Q_T for $h \to 0$. Moreover, the discrete derivatives $\widetilde{\partial_k^+ \psi_h}|_{Q_T}$ converge uniformly to $\partial_{x_k}\psi$. Furthermore, note that, for $\psi \in C_0^\infty(Q_T)$,

$$\begin{aligned} \int_0^T \langle u_l(t),\, \psi_l(t) \rangle_{\mathcal{H}_0^1} \, dt &= \int_0^T \big(u_l(t),\, \psi_l(t)\big)_{\mathcal{L}^2} \, dt \\ &= \int_0^T \big(\tilde{u}_l(t),\, \tilde{\psi}_l(t)\big)_{L^2} \, dt. \end{aligned} \tag{68}$$

The proof of Theorem (3.3) is based on the following auxiliary result that will be shown below.

Theorem 4.5. *Assume that the scaling relations* (25) *are satisfied, and denote by v_l the solutions of the approximating problem* (56) *with $D_l = \frac{1}{2m} l^2 d$ to the initial value $v_{l,0} = u_l(0)$. Then*

$$\sup_{t \le T} E\Big[\|u_l(t) - v_l(t)\|_{\mathcal{H}^{-1}}^2 \Big] \to 0.$$

Proof of Theorem 3.3. Let ψ_l be the approximating lattice function of an arbitrary function $\psi \in C_0^\infty(Q_T)$ as defined above. (Assume that ψ is not identically zero to avoid trivialities.) Then

$$\begin{aligned} &P\left[\left| \int_0^T \int_G u_l \, \psi \, dx \, dt - \int_0^T \int_G u \, \psi \, dx \, dt \right| > \varepsilon \right] \\ &\quad \le P\left[\left| \int_0^T \int_G (u_l - v_l) \, \psi \, dx \, dt \right| > \varepsilon/2 \right] + \\ &\quad\quad + P\left[\left| \int_0^T \int_G (u - v_l) \, \psi \, dx \, dt \right| > \varepsilon/2 \right]. \end{aligned} \tag{69}$$

The second term in the sum vanishes for $l \to 0$ because v_l converges weakly to u in $L^2(0,T;L^2)$. As for the first term, observe that

$$P\left[\left| \int_0^T \int_G (u_l - v_l) \, \psi \, dx \, dt \right| > \varepsilon/2 \right]$$

$$\leq P\left[\left|\int_0^T\int_G (u_l - v_l)\,\psi_l\,dx\,dt\right| > \varepsilon/4\right] + \tag{70}$$
$$+ P\left[\left|\int_0^T\int_G (u_l - v_l)\,(\psi - \psi_l)\,dx\,dt\right| > \varepsilon/4\right].$$

Again the second term in the sum tends to zero for $l \to 0$, since almost surely $\sup_l \|u_l - v_l\|_{L^1(Q_T)} < \infty$ (cf. (18)) and $\|\psi - \psi_l\|_{L^\infty(Q_T)} \to 0$. Let now $C > 0$ be a constant such that $\|\psi_l\|_{L^2(0,T;\mathcal{H}_0^1)} \geq C$ for sufficiently small l. Then

$$P\left[\left|\int_0^T\int_G (u_l - v_l)\,\psi_l\,dx\,dt\right| > \varepsilon/4\right]$$
$$= P\left[\left|\int_0^T \big\langle u_l(t) - v_l(t),\, \psi_l(t)\big\rangle_{\mathcal{H}_0^1}\,dt\right| > \varepsilon/4\right]$$
$$\leq P\left[\left(\int_0^T \|u_l(t) - v_l(t)\|_{\mathcal{H}^{-1}}^2\,dt\right)^{1/2}\left(\int_0^T \|\psi_l(t)\|_{\mathcal{H}_0^1}^2\,dt\right)^{1/2} > \varepsilon/4\right] \tag{71}$$
$$\leq P\left[\left(\int_0^T \|u_l(t) - v_l(t)\|_{\mathcal{H}^{-1}}^2\,dt\right)^{1/2} > \varepsilon/(4C)\right]$$
$$\leq \frac{(4C)^2}{\varepsilon^2}\,E\left[\int_0^T \|u_l(t) - v_l(t)\|_{\mathcal{H}^{-1}}^2\,dt\right] \to 0.$$

□

It remains to prove the auxiliary theorem 4.5. The proof is based on the next lemma that identifies a martingale related to the process $\|u_l(t) - v_l(t)\|_{\mathcal{H}^{-1}}^2$, $t \leq T$.

Lemma 4.6. *The process $(M_l(t))_{t\leq T}$ given by*

$$M_l(t) = \|u_l(t) - v_l(t)\|_{\mathcal{H}^{-1}}^2 - \|u_l(0) - v_l(0)\|_{\mathcal{H}^{-1}}^2 + \tag{72}$$
$$+ 2\int_0^t \big\langle A_l(u_l(s)) - A_l(v_l(s)),\, u_l(s) - v_l(s)\big\rangle_{\mathcal{L}^2}\,ds - R_l(t),$$

where

$$R_l(t) = \frac{1}{n}\int_0^t \big(d(u_l(s))\,u_l(s),\, \beta_l\big)_{\mathcal{L}^2}, \tag{73}$$

is a martingale. Here the lattice functions β_l satisfy $\max_{z\in\mathcal{G}_l} |\beta_l(z)| \leq C$ for a constant C independent of l.

Proof. Consider for fixed $w_l \in \mathcal{L}^2$ the function $g(\cdot, w_l) : S_l \to \mathbb{R}$ given by $u_l \mapsto g(u_l, w_l) = \|u_l - w_l\|_{\mathcal{H}^{-1}}^2$, and recall that

$$\|u_l - w_l\|_{\mathcal{H}^{-1}}^2 = |||u_l - w_l|||_{\mathcal{H}^{-1}}^2 = \big(u_l - w_l,\, (-\Delta_l)^{-1}(u_l - w_l)\big)_{\mathcal{L}^2}. \tag{74}$$

We are going to compute $L_l g(u_l, w_l)$.

$$\begin{aligned}
L_l g(u_l, w_l) = &\sum_{z\in\mathcal{G}_l^1}\sum_{k=1}^{m} n\,\frac{1}{2m} d(u_l(z))\,u_l(z)\times \\
&\times\Big(\big|\big|\big|u_l - \tfrac{1}{n}\chi_z + \tfrac{1}{n}\chi_{(z-le_k)} - w_l\big|\big|\big|^2_{\mathcal{H}^{-1}} - \\
&\quad - 2\,|||u-w|||^2_{\mathcal{H}^{-1}} + \\
&\quad + \big|\big|\big|u_l - \tfrac{1}{n}\chi_z + \tfrac{1}{n}\chi_{(z+le_k)} - w_l\big|\big|\big|^2_{\mathcal{H}^{-1}}\Big) \qquad (75)\\
= &\sum_{z\in\mathcal{G}_l^1}\sum_{k=1}^{m} n\,\frac{1}{2m} d(u_l(z))\,u_l(z)\times \\
&\times\Big(\frac{2}{n}\big(u_l - w_l,\, \chi_{(z-le_k)} - 2\,\chi_z + \chi_{(z+le_k)}\big)_{\mathcal{H}^{-1}} + \\
&\quad + \frac{1}{n^2}\,\big|\big|\big|\chi_{(z-le_k)} - \chi_z\big|\big|\big|^2_{\mathcal{H}^{-1}} + \frac{1}{n^2}\,\big|\big|\big|\chi_{(z+le_k)} - \chi_z\big|\big|\big|^2_{\mathcal{H}^{-1}}\Big).
\end{aligned}$$

We set $\tilde{u}_l = (-\Delta_l)^{-1}u_l$, and $\tilde{w}_l = (-\Delta_l)^{-1}w_l$. Hence, we get

$$\begin{aligned}
L_l g(u_l, w_l) = &\sum_{z\in\mathcal{G}_l^1} n\,\frac{1}{2m} d(u_l(z))\,u_l(z)\times \\
&\times\Big(\frac{2}{n} l^{m+2}\big(\Delta_l\tilde{u}_l(z) - \Delta_l\tilde{w}_l(z)\big) + \frac{1}{n^2}\tilde{\beta}_l(z)\Big),
\end{aligned} \tag{76}$$

where

$$\begin{aligned}
\tilde{\beta}_l(z) = \sum_{k=1}^{m}\Big(&\big(\chi_{(z-le_k)} - \chi_z,\, (-\Delta_l)^{-1}(\chi_{(z-le_k)} - \chi_z)\big)_{\mathcal{L}^2} + \\
&+ \big(\chi_{(z+le_k)} - \chi_z,\, (-\Delta_l)^{-1}(\chi_{(z+le_k)} - \chi_z)\big)_{\mathcal{L}^2}\Big).
\end{aligned} \tag{77}$$

Note that

$$\left|\tilde{\beta}_l(z)\right| \le C\sum_{k=1}^{m}\Big(\|\chi_{(z-l)} - \chi_z\|^2_{\mathcal{L}^2} + \|\chi_{(z+l)} - \chi_z\|^2_{\mathcal{L}^2}\Big) \le C\,m\,l, \tag{78}$$

since the discrete Laplacian, as its continuous analogue, has a bounded inverse. We set $\beta_l = \tilde{\beta}_l/(2\,m\,l)$. Finally, we get

$$\begin{aligned}
L_l g(u_l, w_l) &= -2\,\frac{1}{2m}\,l^2\,\big(d(u_l)\,u_l,\, u_l - w_l\big)_{\mathcal{L}^2} + \frac{1}{n}\big(d(u_l)\,u_l,\, \beta_l\big)_{\mathcal{L}^2} \\
&= -2\,a_l(u_l, u_l - w_l) + \frac{1}{n}\big(d(u_l)\,u_l,\, \beta_l\big)_{\mathcal{L}^2}.
\end{aligned} \tag{79}$$

Consider now for fixed $w_l \in \mathcal{L}^2$ the function $h(\cdot, w_l) : [0,T] \to \mathbb{R}$, $t \mapsto h(t, w_l) = \|w_l - v_l(t)\|^2_{\mathcal{H}^{-1}}$, and observe that

$$h'(t, w_l) = -2\,\big(v_l'(t),\, w_l - v_l(t)\big)_{\mathcal{H}^{-1}} = 2\,a_l(v_l(t), w_l - v_l(t)). \tag{80}$$

Let $\Phi : S_l \times [0,T]$, $(u_l, t) \mapsto \Phi(u_l, t) = \|u_l - v_l(t)\|^2_{\mathcal{H}^{-1}}$. It follows again from Dynkin's formula that the process $(M_l(t))_{t\le T}$ given by

$$
\begin{aligned}
M_l(t) &= \Phi(u_l(t), t) - \Phi(u_l(0), 0) - \\
&\quad - \int_0^t \Big(L_l \Phi(u_l(s), s) + \partial_s \Phi(u_l(s), s) \Big)\, ds \\
&= \|u_l(t) - v_l(t)\|^2_{\mathcal{H}^{-1}} - \|u_l(0) - v_l(0)\|^2_{\mathcal{H}^{-1}} - \\
&\quad - \int_0^t \big(L_l g(u_l(s), v_l(s)) + h'(s, u_l(s)) \big)\, ds
\end{aligned} \tag{81}
$$

is a martingale. (Here we do not have to worry about Φ being unbounded, since the particle density process is, for fixed l, bounded by construction.) Substituting the explicit computations in the equation above yields (72). □

Proof of Theorem 4.5. We set $\hat{d} = \sup_{\mathbb{R}} d$. By taking expectations in the martingale formula (72) and making use of the monotonicity of A_l we get the estimate

$$
\begin{aligned}
E\Big[\|u_l(t) - v_l(t)\|^2_{\mathcal{H}^{-1}} \Big] &\le \|u_l(0) - v_l(0)\|^2_{\mathcal{H}^{-1}} + \\
&\quad + C\, E \int_0^t \frac{\hat{d}}{n} \big(u_l(s),\, 1 \big)_{\mathcal{L}^2}\, ds \\
&\le \|u_l(0) - v_l(0)\|^2_{\mathcal{H}^{-1}} + C\, \frac{\hat{d}}{n}\, \|u_l(0)\|_{\mathcal{L}^2} \to 0.
\end{aligned} \tag{82}
$$

Here the second inequality is due to the estimate (18). □

4.3. **Gradient-activated diffusion.**

4.3.1. *An auxiliary problem.* The approximating problem on the interior lattice points $\mathcal{G}_h$ is given by the following system of ODEs:

$$
\begin{cases}
u_h' - \displaystyle\sum_{k=1}^m \partial_k^- D_h(\partial_k^+ u_h)\, \partial_k^+ u_h = 0 & \text{in } \mathcal{G}_h^1 \times (0,T) \\
u_h = 0 & \text{on } (\mathcal{G}_h \setminus \mathcal{G}_h^1) \times [0,T] \\
u_h(\cdot, 0) = u_{h,0} & \text{in } \mathcal{G}_h^1,
\end{cases} \tag{83}
$$

where the function $D_h : \mathbb{R} \to \mathbb{R}_0^+$ is assumed to satisfy conditions (22a) and (22b). Moreover, we assume that $\sup_{\mathbb{R}} |D_h - D| \to 0$ for $h \to 0$. This is again a finite-dimensional ODE system with continuous right-hand side, and it has a local solution according to the Peano theorem. The existence of a solution on the entire interval $[0,T]$ follows from the derivation of the a-priori estimate (87) below. We define the mapping $a_h : \mathcal{H}_0^1 \times \mathcal{H}_0^1 \to \mathbb{R}$ by

$$
a_h(u_h, v_h) = \sum_{k=1}^m \big(D_h(\partial_k^+ u_h)\, \partial_k^+ u_h,\, \partial_k^+ v_h \big)_{\mathcal{L}^2}, \quad u_h, v_h \in \mathcal{H}_0^1. \tag{84}
$$

Note that $a_h(\cdot,\cdot)$ once more induces a bounded monotone operator $A_h : \mathcal{H}_0^1 \to \mathcal{H}^{-1}$ by

$$\big\langle A_h(u_h),\, v_h \big\rangle_{\mathcal{H}_0^1} = a_h(u_h, v_h), \quad u_h, v_h \in \mathcal{H}_0^1. \tag{85}$$

The solution of (83) can be regarded as a function in $C^1([0,T], \mathcal{H}_0^1)$ that solves the discrete weak problem

$$\frac{d}{dt}\big(u_h(t),\, v_h\big)_{\mathcal{L}^2} + a_h(u_h(t), v_h) = 0 \tag{86}$$

for all $v_h \in \mathcal{H}_0^1$ and $t \in [0,T]$.

Here, as in Section 4.1, we first show strong convergence of the solutions of the approximating problem (83) in $L^2(0,T;L^2)$.

Theorem 4.7. *Let u be the solution of the weak PDE problem* (12) *to the initial value u_0, and let (u_h), $h \searrow 0$, be a sequence of solutions of the approximating problem* (83) *to the initial value $u_{h,0}$. If $u_{h,0}$ converges strongly to u_0 in L^2, then u_h converges strongly to u in $L^2(0,T;L^2)$.*

Sketch of proof. The a-priori estimates

$$\sup_h \max_{0\le t\le T} \|u_h(t)\|_{\mathcal{L}^2} < \infty, \tag{87}$$

$$\sup_h \|u_h\|_{L^2(0,T;\mathcal{H}_0^1)} < \infty, \tag{88}$$

$$\sup_h \|u_h'\|_{L^2(0,T;\mathcal{H}^{-1})} < \infty, \tag{89}$$

follow from the weak formulation of the approximating problem by inserting $u_h(t)$ for v_h and integrating over time. The passage to the limit of a weakly convergent subsequence in $L^2(0,T;L^2)$ can again be carried out with techniques from Temam (2001) and the Minty lemma. Strong convergence follows from a discrete analogue of the Aubin-Lions compactness theorem. For more details we refer again to Reichert (2006). □

4.3.2. *Convergence of the particle density.* From now on v_l denotes the solution of the approximating problem (26) and u_l the stochastic particle density. In view of Theorem 4.7, the law of large numbers is an immediate consequence of the following result.

Theorem 4.8. *Let u be the solution of the weak problem* (30) *to the initial value u_0. Assume that the scaling relations* (31) *are satisfied, and denote by v_l the solutions of the approximating problem* (83) *with $D_l = l^2\, d$ to the initial value $v_{l,0} = u_l(0)$. If $u_l(0)$ converges strongly to u_0 in L^2, then*

$$\sup_{t\le T} E\Big[\|u_l(t) - v_l(t)\|_{\mathcal{L}^2}^2\Big] \to 0.$$

The proof of the above theorem rests again on a lemma that identifies a related martingale.

Lemma 4.9. *The process* $(M_l(t))_{t\leq T}$ *given by*

$$M_l(t) = \|u_l(t) - v_l(t)\|_{\mathcal{L}^2}^2 - \|u_l(0) - v_l(0)\|_{\mathcal{L}^2}^2 + \\ + \int_0^t \big\langle A_l(u_l(s)) - A_l(v_l(s)),\, u_l(s) - v_l(s)\big\rangle_{\mathcal{H}_0^1}\, ds - R_l(t), \tag{90}$$

where

$$R_l(t) = \frac{2}{n}\sum_{k=1}^m \int_0^t \big((d(\partial_k^+ u_l(s)) + d(\partial_k^- u_l(s)))\, u_l(s),\, 1\big)_{\mathcal{L}^2}\, ds, \tag{91}$$

is a martingale.

Proof. Consider for fixed $w_l \in \mathcal{H}_0^1$ the function $g(\cdot, w_l) : S_l \to \mathbb{R}$ given by $u_l \mapsto g(u_l, w_l) = \|u_l - w_l\|_{\mathcal{L}^2}^2$. We compute $L_l g(u_l, w_l)$.

$$\begin{aligned}
L_l g(u_l, w_l) &= \sum_{z\in\mathcal{G}_l^1}\sum_{k=1}^m n\, d(\partial_k^+ u_l(z))\, u_l(z)\times \\
&\quad \Big(\|u_l - \tfrac{1}{n}\chi_z + \tfrac{1}{n}\chi_{(z+le_k)} - w_l\|_{\mathcal{L}^2}^2 - \|u_l - w_l\|_{\mathcal{L}^2}^2\Big) + \\
&\quad + \sum_{z\in\mathcal{G}_l^1}\sum_{k=1}^m n\, d(-\partial_k^- u_l(z))\, u_l(z)\times \\
&\quad \times \Big(\|u_l - \tfrac{1}{n}\chi_z + \tfrac{1}{n}\chi_{(z-le_k)} - w_l\|_{\mathcal{L}^2}^2 - \|u_l - w_l\|_{\mathcal{L}^2}^2\Big) \\
&= \sum_{z\in\mathcal{G}_l^1}\sum_{k=1}^m n\, d(\partial_k^+ u_l(z))\, u_l(z)\times \\
&\quad \times \left(\frac{2}{n}\big(u_l - w_l,\, \chi_{(z+le_k)} - \chi_z\big)_{\mathcal{L}^2} + \frac{1}{n^2}\|\chi_{(z+le_k)} - \chi_z\|_{\mathcal{L}^2}^2\right) + \\
&\quad + \sum_{z\in\mathcal{G}_l^1}\sum_{k=1}^m n\, d(-\partial_k^+ u_l(z - le_k))\, u_l(z)\times \\
&\quad \times \left(\frac{2}{n}\big(u_l - w_l,\, \chi_{(z-le_k)} - \chi_z\big)_{\mathcal{L}^2} + \frac{1}{n^2}\|\chi_{(z-le_k)} - \chi_z\|_{\mathcal{L}^2}^2\right) \\
&= \sum_{z\in\mathcal{G}_l^1}\sum_{k=1}^m n\, d(\partial_k^+ u_l(z))\, u_l(z)\times \\
&\quad \times \left(2\frac{l^m}{n}\Big(u_l(z + le_k) - w_l(z + le_k) - \big(u_l(z) - w_l(z)\big)\Big)\right) + \\
&\quad + \sum_{z\in\mathcal{G}_l^1}\sum_{k=1}^m n\, d(-\partial_k^+ u_l(z - le_k))\, u_l(z)\times \\
&\quad \times \left(2\frac{l^m}{n}\Big(u_l(z - le_k) - w_l(z - le_k) - \big(u_l(z) - w_l(z)\big)\Big)\right)
\end{aligned} \tag{92}$$

$$+\sum_{z\in\mathcal{G}_l^1}\sum_{k=1}^{m} n\, u_l(z)\, 2\frac{l}{n^2}\Big(d(\partial_k^+ u_l(z)) + d(-\partial_k^- u_l(z))\Big).$$

By introducing discrete derivatives and making use of assumption (22b) it follows that

$$\begin{aligned} L_l g(u_l, w_l) &= 2\sum_{z\in\mathcal{G}_l^1}\sum_{k=1}^{m} l^{m+1}\, d(\partial_k^+ u_l(z))\, u_l(z)\,\big(\partial_k^+ u_l(z) - \partial_k^+ w_l(z)\big)- \\ &\quad - 2\sum_{z\in\mathcal{G}_l^1}\sum_{k=1}^{m} l^{m+1}\, d(\partial_k^+ u_l(z - le_k))\, u_l(z)\times \\ &\qquad \times\big(\partial_k^+ u_l(z-le_k) - \partial_k^+ w_l(z-le_k)\big)+ \\ &\quad + 2\sum_{z\in\mathcal{G}_l^1}\sum_{k=1}^{m} \frac{l^m}{n}\, u_l(z)\,\big(d(\partial_k^+ u_l(z)) + d(\partial_k^- u_l(z))\big) \\ &= 2\, l^m \sum_{z\in\mathcal{G}_l^1}\sum_{k=1}^{m} u_l(z)\,\partial_k^-\big(l^2 d(\partial_k^+ u_l)(\partial_k^+ u_l - \partial_k^+ w_l)\big)(z)+ \\ &\quad + 2\sum_{z\in\mathcal{G}_l^1}\sum_{k=1}^{m} \frac{l^m}{n}\, u_l(z)\,\big(d(\partial_k^+ u_l(z)) + d(\partial_k^- u_l(z))\big). \end{aligned} \tag{93}$$

Hence, by a discrete integration by parts,

$$\begin{aligned} L_l g(u_l, w_l) &= -2\sum_{k=1}^{m}\big(l^2\, d(\partial_k^+ u_l)\,\partial_k^+ u_l,\ \partial_k^+ u_l - \partial_k^+ w_l\big)_{\mathcal{L}^2}+ \\ &\quad + \frac{2}{n}\sum_{k=1}^{m}\big(\big(d(\partial_k^+ u_l) + d(\partial_k^- u_l)\big)\, u_l,\ 1\big)_{\mathcal{L}^2} \\ &= -2\, a_l(u_l, u_l - w_l) + \frac{2}{n}\sum_{k=1}^{m}\big((d(\partial_k^+ u_l) + d(\partial_k^- u_l))\, u_l,\ 1\big)_{\mathcal{L}^2}. \end{aligned} \tag{94}$$

We now consider for arbitrary but fixed $w_l \in \mathcal{H}_0^1$ the function $h(\cdot, w_l) : [0,T] \to \mathbb{R}$, $t \mapsto h(t, w_l) = \|w_l - v_l(t)\|_{\mathcal{L}^2}^2$. Note that

$$h'(t, w_l) = -2\big(v_l'(t),\ w_l - v_l(t)\big)_{\mathcal{L}^2} = 2\, a_l(v_l(t), w_l - v_l(t)). \tag{95}$$

Let $\Phi : S_l \times [0,T] \to \mathbb{R}$ be given by $(u_l, t) \mapsto \Phi(u_l, t) = \|u_l(t) - v_l(t)\|_{\mathcal{L}^2}^2$. It follows again from Dynkin's formula that the process $(M_l(t))_{t\le T}$ defined by

$$\begin{aligned} M_l(t) &= \Phi(u_l(t), t) - \Phi(u_l(0), 0)- \\ &\quad - \int_0^t \Big(L_l\Phi(u_l(s), s) + \partial_s\Phi(u_l(s), s)\Big)\, ds \\ &= \|u_l(t) - v_l(t)\|_{\mathcal{L}^2}^2 - \|u_l(0) - v_l(0)\|_{\mathcal{L}^2}^2- \end{aligned} \tag{96}$$

$$- \int_0^t \Big(L_l g(u_l(s), v_l(s)) + h'(s, u_l(s)) \Big) \, ds$$

is a martingale. Substituting the explicit computations above yields (90). □

Proof of Theorem 4.8. By taking expectations in Eq. (90) and making use of the monotonicity of A_l we get the estimate

$$\begin{aligned} E\Big[\|u_l(t) - v_l(t)\|_{\mathcal{L}^2} \Big] &\leq \|u_l(0) - v_l(0)\|_{\mathcal{L}^2} + \\ &\quad + 2\, mE \int_0^t \frac{\hat{d}}{n} \big(u_l(s),\, 1\big)_{\mathcal{L}^2} \, ds \\ &\leq \|u_l(0) - v_l(0)\|_{\mathcal{L}^2} + C\, \frac{\hat{d}}{n} \|u_l(0)\|_{\mathcal{L}^2}, \end{aligned} \tag{97}$$

where $\hat{d} = \sup_{\mathbb{R}} d$. In view of the hypotheses, we can conclude that the right hand side tends to zero, which finishes the proof. □

5. Discussion

We have seen that by using roughly the same procedure it is possible to derive laws of large numbers for several typical instances of the general mesoscopic stochastic particle model introduced in Section 2.2. While the scaling relations (13a), (13b) and the corresponding relations for the other models appear natural, condition (13c) is more difficult to justify in physical terms. It serves to damp out the fluctuating term in the remainder $R_l(t)$ (Eq. (44)) that stems from diffusive jumps. Stated in terms of n and l, condition (13c) reads $(1/n)/l^2 \to 0$. Heuristically, $1/\sqrt{n}$ is a measure for the size of fluctuations of the particle densities. Therefore $(1/\sqrt{n})/l$ may be interpreted as a measure for the gradients of the particle densities caused by fluctuations. Condition (13c) forces these gradients to vanish asymptotically.

The scaling relation (13c) also appears in Arnold & Theodosopulu (1980) and Kotelenez (1986) in their treatment of single-species models with linear reaction kinetics. In addition, Kotelenez (1986, 1988) is able to prove a law of large numbers in a weak norm for a single-species model with linear or polynomial kinetics using only (13a) and (13b). Under the same hypotheses Blount (1994) has a stronger result for the model with polynomial kinetics. In addition Blount (1994) discusses a law of large numbers for a particular model where n is kept constant while l goes to zero. However, all authors mentioned above work with particle densities defined on the unit cube in $\mathbb{R}^m$, which has the advantage that the eigenvalues and eigenfunctions of the Laplacian are explicitly known. This knowledge is exploited in Blount (1994) to get rid of condition (13c). Different time scales for the chemical reactions are treated in Ball et al. (2006) for some spatially homogeneous models.

A motivation for considering weak solutions of the limit equations is that the nonlinear parabolic PDEs that appear as limit dynamics of stochastic particle models might have solutions that are not differentiable in the classical sense, especially if a nonlinear diffusion operator is involved. If, on the other hand, the solution of the limit equations is sufficiently smooth, as is the case, e.g., for the model with linear diffusion and Lipschitz continuous reaction rates (in case of sufficiently smooth data), one can obtain explicit rates for the convergence of the solution $\boldsymbol{v}_l$ of the auxiliary problem to the solution of the limit equation $\boldsymbol{u}$. Let us assume that $\|\boldsymbol{u} - \boldsymbol{v}_l\|_{L^2(0,T;\boldsymbol{L}^2)} = O(l)$, and suppose that $n = O(l^{-\alpha})$ and $\hat{d} = O(l^{-\beta})$, where $\alpha > \beta > 0$, in order to satisfy condition (13c). Moreover, assume that $\boldsymbol{u}_l(0) = \boldsymbol{v}_{l,0}$ and $\sup_{\mathbb{R}^{n_s}} |\boldsymbol{f}_l - \boldsymbol{f}| = O(l)$. Then it can easily be seen from the estimate (54) that the rate of convergence in the law of large numbers is $O(l^{\alpha-\beta})$ if $\beta + 1 > \alpha > \beta$, and $O(l)$ if $\alpha \geq \beta + 1$. In other words, the rate of convergence is determined by the ratio $\hat{d}/n$ if $\alpha - \beta < 1$.

Although we were able to handle quite general classes of reaction-diffusion systems, the examples considered in the present work are by no means exhaustive. Nevertheless, similar techniques might be applied to models that include convection, cross-diffusion of different species, or ‘freezing’ of particles (Stefan problems). The essential limiting condition seems to be the monotonicity of the diffusion operator and its discrete analogue.

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